Application No. 10/522,839

Reply to Office Action of August 18, 2008

AMENDMENTS TO THE CLAIMS

Docket No.: 15588-00007-US

- 1. (Currently Amended) A proton-conducting polymer membrane comprising polymers containing phosphonic acid groups which is obtained by a process comprising the steps
- A) mixing of mixing at least 20% by weight of based on the total weight of the mixture of vinyl-containing phosphonic acid with one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids, esters thereof, acid halides thereof or anhydrides thereof which contain at least two acid groups per carboxylic acid monomer, and/or mixing of vinyl-containing phosphonic acid with one or more aromatic and/or heteroaromatic diamino carboxylic acids, esters thereof, acid halides thereof or anhydrides thereof,
- B) heating of the mixture obtainable according to step A) under inert gas at temperatures of up to 350°C to form polyazole polymers a polyazole polymer,
- C) application of a layer using the mixture from step A) and/or B) to a support,
- D) polymerization of the vinyl-containing phosphonic acid present in the sheet-like structure obtainable according to step C)

and wherein the membrane has a proton conductivity of at least 0.001 S/cm @ 120 °C.

- 2. (Original) The membrane as claimed in claim 1, characterized in that 3,3',4,4'tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine, 1,2,4,5-tetraaminobenzene, bis(3,4diaminophenyl) sulfone, bis(3,4-diaminophenyl) ether, 3,3',4,4'-tetraaminobenzophenone, 3,3',4,4'-tetraaminodiphenylmethane and 3,3',4,4'-tetraaminodiphenyldimethylmethane are used as aromatic tetraamino compounds.
- 3. (Previously presented) The membrane as claimed in claim 1, characterized in that isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,Ndimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic acid, 2,3-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic

acid, tetrafluoroterephthalic acid,1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, bis(4-carboxyphenyl) ether, benzophenone-4,4'-dicarboxylic acid, bis(4-carboxyphenyl) sulfone, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl)hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-carboxycinnamic acid, or C1-C20-alkyl esters or C5-C12-aryl esters thereof, or anhydrides thereof or acid chlorides thereof are used as aromatic carboxylic acids.

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- 4. (Previously presented) The membrane as claimed in claim 1, characterized in that tricarboxylic acids, C1-C20-alkyl esters thereof, C5-C12-aryl esters thereof, anhydrides thereof or acid chlorides thereof or tetracarboxylic acids, C1-C20-alkyl esters thereof, C5-C12-aryl esters thereof, anhydrides thereof or acid chlorides thereof are used as aromatic carboxylic acids.
- 5. (Original)The membrane as claimed in claim 4, characterized in that 1,3,5-benzenetricarboxylic acid (trimesic acid); 2,4,5-benzenetricarboxylic acid (trimellitic acid); (2-carboxyphenyl)iminodiacetic acid, 3,5,3'-biphenyltricarboxylic acid; 3,5,4'-biphenyltricarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzene-1,2,4,5-tetracarboxylic acid; naphthalene-1,4,5,8-tetracarboxylic acid, 3,5,3',5'-biphenyltetracarboxylic acid, benzophenonetetracarboxylic acid, 3,3',4,4'-biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxylic acid and/or 1,4,5,8-naphthalenetetracarboxylic acid are used as aromatic carboxylic acids.
- 6. (Previously presented) The membrane as claimed in claim 4, characterized in that the content of tricarboxylic acids and/or tetracarboxylic acids is in the range from 0 to 30 mol% based on dicarboxylic acid used.
- 7. (Original) The membrane as claimed in claim 1, characterized in that heteroaromatic dicarboxylic acids, heteroaromatic tricarboxylic acids and/or heteroaromatic tetracarboxylic acids containing at least one nitrogen, oxygen, sulfur or phosphorus atom in the aromatic are used as heteroaromatic carboxylic acids.

8. (Original) The membrane as claimed in claim 7, characterized in that pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid, and also C1-C20-alkyl esters or C5-C12-aryl esters thereof, or anhydrides thereof or acid chlorides thereof are used.

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- 9. (Original) The membrane as claimed in claim 1, characterized in that diaminobenzoic acid and/or monohydrochloride and dihydrochloride derivatives thereof are used as aromatic diamino carboxylic acids.
- 10. (Previously presented) The membrane as claimed in claim 1, characterized in that the mixture prepared in step A) and/or step B) comprises compounds of the formula

$$R - (PO_3Z_2)_x$$

where

R is a bond, a C1-C15-alkyl group, C1-C15-alkoxy group, ethylenoxy group or C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, COOZ, -CN, or NZ₂,

the radicals Z are each, independently of one another, hydrogen, a C1-C15-alkyl group, C1-C15-alkoxy group, ethylenoxy group or C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, or -CN, and

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and/or of the formula

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where

R is a bond, a C1-C15-alkyl group, C1-C15-alkoxy group, ethylenoxy group or C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, COOZ-CN, NZ₂ -CN, or NZ₂,

the radicals Z are each, independently of one another, hydrogen, a C1-C15-alkyl group, C1-C15-alkoxy group, ethylenoxy group or C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, or -CN, and

x is 1, 2, 3, 4, 5, 6, 7, 8, 9 or 10, and/or of the formula

where

A is a group of the formulae $COOR^2$, CN, $CONR^2_2$, OR^2 and/or R^2 , where R^2 is hydrogen, a C1-C15-alkyl group, C1-C15-alkoxy group, ethylenoxy group or C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, COOZ, -CN, or NZ_2 ,

R is a bond, a divalent C1-C15-alkylene group, divalent C1-C15-alkylenoxy group, for example ethylenoxy group, or divalent C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, COOZ, -CN, or NZ₂,

the radicals Z are each, independently of one another, hydrogen, a C1-C15-alkyl group, C1-C15-alkoxy group, ethylenoxy group or C5-C20-aryl or heteroaryl group, with the above radicals themselves being able to be substituted by halogen, -OH, or -CN, and

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(Original)The membrane as claimed in claim 1, characterized in that monomers which 11. are capable of effecting crosslinking and have at least 2 carbon-carbon double bonds are polymerized in step D).

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- 12. (Original) The membrane as claimed in claim 1, characterized in that the polymerization in step D) is brought about by substance capable of forming free radicals.
- 13. (Original) The membrane as claimed in claim 1, characterized in that the polymerization in step D) is effected by irradiation with IR or NIR light, UV light, \(\beta\)-rays, \(\gamma\)- rays and/or electron beams.
- (Original) The membrane as claimed in claim 1, characterized in that the mixture 14. produced in step A) and/or step B) comprises dissolved, dispersed and/or suspended polymer.
- 15. (Previously presented) The membrane as claimed in claim 1, characterized in that a layer having a thickness of from 20 to 4000 µm is produced in step C).
- 16. (Previously presented) The membrane as claimed in claim 1, characterized in that the membrane formed in step D) has a thickness in the range from 15 to 3000 µm.
- 17-18 (Cancelled)
- 19. (Withdrawn) A membrane-electrode unit comprising at least one electrode and at least one membrane as claimed in claim 1.
- (Cancelled) 20.
- 21. (Withdrawn) A fuel cell comprising one or more membrane-electrode units as claimed in claim 19.

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22. (Withdrawn) A process for producing proton-conducting polymer membranes comprising polymers containing phosphonic acid groups, which comprises the steps

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- A) mixing of one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids, esters thereof, acid halides thereof or anhydrides thereof which contain at least two acid groups per carboxylic acid monomer or mixing of one or more aromatic and/or heteroaromatic diamino carboxylic acids, esters thereof, acid halides thereof or anhydrides thereof with vinyl-containing phosphonic acid,
- B) heating of the mixture obtainable according to step A) under inert gas at temperatures of up to 350°C to form polyazole polymers,
- C) application of a layer using the mixture from step A) and/or B) to a support,
- D) polymerization of the vinyl-containing phosphonic acid.
- 23. (Previously presented) The membrane as claimed in claim 4, characterized in that the content of tricarboxylic acids and/or tetracarboxylic acids is in the range 0.5 to 10 mol%, based on dicarboxylic acid used.
- 24. (New) The membrane as claimed in claim 1, wherein the membrane has a proton conductivity of at least 10 S/cm @ 120 °C.
- 25. (New) The membrane as claimed in claim 1, wherein the membrane has a proton conductivity of at least 20 S/cm @ 120 °C.
- 26. (New) The membrane as claimed in claim 1, wherein the vinyl-containing phosphonic acid is present in step A) in an amount of at least 30% by weight of based on the total weight of the mixture.
- 27. (New) The membrane as claimed in claim 1, wherein the vinyl-containing phosphonic acid is present in step A) in an amount of at least 50% by weight of based on the total weight of the mixture.

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28. (New) The membrane as claimed in claim 1, wherein the polyzole polymer formed in step B) has recurring azole units in the polymer greater than or equal to 10.

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- 29. (New) The membrane as claimed in claim 1, wherein the polyzole polymer formed in step B) has recurring azole units in the polymer greater than or equal to 100.
- 30. (New) The membrane as claimed in claim 25, wherein the polyzole polymer formed in step B) has recurring azole units in the polymer greater than or equal to 100 and wherein the vinyl-containing phosphonic acid is present in step A) in an amount of at least 50% by weight of based on the total weight of the mixture.

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